Short Communication _____ *Thematic Section:* Theoretical Research.
Reference Object Identifier – ROI: jbc-02/17-50-6-30 *Subsection:* Physical Chemistry.
Publication is available for discussion in the framework of the on-line Internet conference "*Butlerov readings*".
http://butlerov.com/readings/
Submitted on June 3, 2017.

Chaotic oscillations in a simple heterogeneous catalytic reaction

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Keywords*:* heterogeneous catalytic reaction, mechanism, stage scheme, law of acting masses, nonstationary kinetics, chaotic oscillations.

Abstract

For heterogeneous catalytic reactions irregular and chaotic oscillations of reagent concentrations and reaction rates in time have been experimentally observed for heterogeneous catalytic reactions of hydrogen oxidation, carbon monoxide and other reactions. Various approaches and mechanisms are used, applying more complicated assumptions: different various variants of nonideal kinetics different from the law of effective masses; mechanisms of multicenter adsorption involving several types of active catalyst centers; mechanisms including "buffer" stages; mechanisms taking into account the interaction of substances in the adsorbed state and their dissolution in the near-surface layer of the catalyst; parallel reaction of different catalytic oscillators on different types of catalyst centers (superposition of oscillations); mechanisms that allow for random perturbations (noise-induced chaos); phenomenological models using non-standard kinetic laws and unusual model constructions ("reservoirs"); various combinations of these approaches to describe chaotic oscillations in heterogeneous catalytic reactions. Thus, nowadays no mechanisms have been found so far that can describe chaotic oscillations of heterogeneous catalytic reactions in gradient chemical reactors within the framework of models of an ideal adsorbed layer and the law of acting masses. Due to this it is necessary to describe the complex dynamic behavior of heterogeneous catalytic reactions by simple mechanisms without complicating assumptions. A special role in the origin of oscillatory processes is played by autocatalytic stages capable of generating unstable regimes in simple dynamical systems with minimal molecularity. This article presents a simple four-stage scheme with two autocatalytic stages and establishes the possibility of describing it with the help of its chaotic oscillations in heterogeneous catalytic reactions within the framework of an ideal adsorbed layer and the law of acting masses. The randomness of oscillations is confirmed by numerical calculations of the kinetic model and the Lyapunov's exponent.

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